

Docket No. BFGRP0313USB

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re **PATENT** application of:

Appellants: John W. Robinson et al.

Application No.: 10/612,850

For: **RUBBER TOUGHENED EPOXY RESINS FOR RESIN
INFUSION PROCESSING, METHODS OF USING THE
RESINS AND ARTICLES MADE THEREFROM**

Filing Date: July 3, 2003

Examiner: Robert E. Sellers

Art Unit: 1796

Confirmation No.: 6883

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Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPELLANT'S REPLY BRIEF

Dear Sir:

This paper is Appellant's Reply Brief in response to the Examiner's Answer mailed October 11, 2007 for which a two month period of response was provided. Accordingly, this Reply Brief is timely filed. Kindly enter the following remarks. Appellant respectfully requests reconsideration of the application, reversal of the claims rejections, and allowance of the pending claims.

REMARKS

This Reply Brief is directed to the comments and arguments in the Response to the Argument section in the Examiner's Answer.

1. The Rejection of Claims 1-3, 8-9, 11, and 30 Under 35 U.S.C. 112, First Paragraph, As Failing To Comply With The Enablement Requirement.

Examiner's Answer

The Examiner argues that the basis in fact for the lack of enablement of the disclosed monoepoxide species is the definition provided on page 5, line 29 to page 6, line 1, and various definitions from other sources. The Examiner also stated that the attempt to mitigate the definition in the Handbook of Epoxy Resins does not discount the requirement for the presence of more than one epoxy group. Additionally, the Examiner attempted to support his argument by specifically addressing the Undue Experimentation Factors.

Reply

While the Examiner reiterated his arguments and presented (for the first time) arguments addressing the Undue Experimentation Factors, the Examiner's argument still does not establish that the claims are not enabled.

The Examiner continues to ignore the teachings in the specification and fails to give the claims their broadest reasonable interpretation that is consistent with the specification. When the specification is considered as a whole, the specification does not require an epoxy resin to have more than one epoxy group. Rather, the specification provides that "a host of commercially available resins...can be used in this invention" and that epoxy resins that are "readily available include octadecylene oxide, epichlorohydrin, styrene oxide, vinylcyclohexene oxide, glycidyl methacrylate..." (Page 6, lines 29-30 to page 7, line 1.) Thus, the specification clearly indicates that Appellant considers compounds with a single epoxide group to be an epoxy resin.

Additionally, contrary to the Examiner's arguments, a person skilled in the art would not have understood that an epoxy resin must have two epoxy groups to be curable. Rather, as Appellant has previously discussed, a person skilled in the art would recognize that a network could be formed by reacting a mono-epoxy with a curing agent. The specification states that the curing agent may be any known curing agent including amines, acids, alcohols, and anhydrides. A person skilled in the art would recognize that a network could be formed using a multifunctional curing agent such as, for example, diamines or multifunctional amines, which are known in the art.¹ Further, the specification discloses that the curing agent may be a dianhydride, which also provides multiple reactive sites to react with an epoxy group.

A person skilled in the art would also understand that certain curing agents may provide a single reaction site that is capable of reacting with more than one epoxide group and which could react with epoxy groups on different molecules to form a network. For example, a primary amine may react with more than one epoxide group. The Handbook of Adhesives shows a reaction path for a primary amine curing agent where the amine group reacts with an epoxy group on a first molecule and then reacts again with an epoxy group on a second molecule to form a completely cross linked system.² Thus, as this reaction path illustrates, a network could be formed by a first monoepoxide molecule reacting with a primary amine molecule and a second monoepoxide molecule reacting again with the same primary amine molecule. Therefore, for these reasons, a person skilled in the art would understand that a monoepoxide may be curable.

The specification also teaches how to make and use the invention. Specifically, the specification discloses that the curable composition (Part A) may be prepared by combining components (a), (b), and (c), and the composition may

¹ See Exhibit E: Handbook of Adhesives, Third Edition (1990) page 354, Table 3 (identifying diethylene triamine and triethylene triamine as current agents). Appellant's Appeal Brief included four Exhibits (A-D), and, to avoid confusion, Appellant's continue this identification sequence in this Reply.

² See Exhibit E, page 351.

be cured by combining the curable composition (Part A) with the curing agent (Part B). (Specification, page 17, lines 8-11.)

Thus, given that (i) the Application shows how to make and cure a curable composition by combining the curable composition and a curing agent, and (ii) a person skilled in the art would understand that an epoxy resin does not require two or more epoxide groups to be curable and that a network could be formed with a curing agent having a reactive group that could react with multiple molecules, the specification provides suitable guidance for a person skilled in the art to cure a composition that includes an epoxy resin with a single epoxy group without the need for undue experimentation.

The Examiner also ignores the fact that the curable composition recited in claim 1 includes a component (c) that is the reaction product of an epoxy resin and a reactive liquid polymer. The epoxy resin of (c) comprises a diglycidyl ether of a bisphenol compound, which is a compound that includes at least two epoxy groups. A person skilled in the art would recognize that an epoxy resin with a single epoxy group could be part of a network, where the epoxy resin with the single epoxy group and an epoxy group from the reaction product (c) react with a multifunctional curing agent or a curing agent capable of reacting with two epoxide groups (e.g., a primary amine).

The Examiner attempted to apply the Undue Experimentation Factors (MPEP § 2164.01(a).) to show a lack of enablement. The Examiner's arguments fail for the reasons described above. Appellants also address each of these arguments in the table below:

| Factor | Examiner's Argument | Appellants Response |
|---------------------------|---|--|
| (A) Breadth of the claims | Breadth denotes epoxy resin; person skilled in the art could not make and use if epoxy resin includes the monoepoxide species because they are not curable. | Disagree: A network may be formed with monoepoxide species and a multifunctional curing agent (e.g., dianhydrides or di-, tri-, or tetra-amines) or a curing agent capable of reacting with two epoxide groups (e.g., a primary amine reacting with an |

| Factor | Examiner's Argument | Appellants Response |
|--|---|---|
| (B) Nature of the invention (C) State of the prior art | One skilled in the art would have known that an epoxy resin must contain more than one epoxy group per molecule for it to be curable as established by documented sources and appellants' own definition, and monoepoxide species set forth in the specification contradict that understanding. | epoxide from different monoepoxide molecules). Disagree: Appellants own specification does not limit epoxy resin to compounds with more than one epoxy group. The Examiner has not shown that a person skilled in the art would not recognize that a network could be formed with an epoxy resin and a multifunctional curing agent or a curing agent capable of reacting with more than one epoxy group. Appellant's arguments demonstrate that a person skilled in the art would recognize that a monoepoxide could be curable. |
| (D) Level of ordinary skill | Level of skill is established by the sources for the definition, appellants own definition, and the cited prior art, all of which confirm that an epoxy resin necessitates the presence of more than one epoxy group. | Disagree: The Examiner narrowly interpreted the definitions from the various sources. The Examiner also ignores (i) the teachings of the specification, which discloses that monoepoxide compounds are considered to be within the scope of epoxy resins, and (ii) the arguments that monoepoxides can be cured. |
| (E) Level of predictability (F) Amount of direction provided (G) Existence of working examples | It cannot be anticipated that the monoepoxides can participate in a cure due to the presence of only one epoxy group per molecule. The only working examples employ epoxy resins with at least two epoxy groups. There are no examples with monoepoxide species to establish their curability. | Disagree: As described above and reiterated in Appellants response to factors (A-C), a person skilled in the art could readily appreciate and select the appropriate curing agent to effect curing of compounds with a monoepoxide. Additionally, the Examiner's arguments ignore the fact that the composition includes a compound (in component (c)) that includes more than one epoxy group. The lack of a specific example with a monoepoxide molecule is not dispositive. The specification provides sufficient guidance in view of the knowledge of persons skilled in the art. |

| Factor | Examiner's Argument | Appellants Response |
|--|---|---|
| (H) Quantity of experimentation needed | The monoepoxides embrace structurally distinct species that would require numerous burdensome experiments to determine wither the claimed curable composition could be formulated and utilized therewith. | The fact that the monoepoxides may embrace structurally distinct species does not mean that numerous burdensome experiments would be needed. The different species share one feature that is relevant to the claim: they have an epoxy group. And, as Appellants have demonstrated in the Appeal Brief and this reply, a person skilled in the art would understand how to select a suitable curing agent to provide a curable composition with a monoepoxide compound. The specification discloses numerous curing agents and others would be known in the art. Even if numerous experiments were required, that does not mean that the experimentation would be an undue burden. A considerable amount of experimentation is permissible if it is merely routine. Since the method of making the composition simply requires combining the curable composition with the curing agent, and a person skilled in the art would recognize what would be required to cure a monoepoxide compound, undue experimentation would not be required to practice the invention. |

Accordingly, the Examiner has not met the burden required under 35 U.S.C. § 112, first paragraph to establish a lack of enablement, and the rejections of claims 1-3, 8-9, 11, and 30 should be reversed.

2. **The Rejection of Claims 1-3, 8-9, 11, and 30 Under 35 U.S.C. § 103(a) As Being Unpatentable over Minamisawa et al. U.S. Patent No. 4,500,660 and Japanese Patent No. 64-60679.**

Examiner's Answer

The Examiner stated that the claimed composition characterizes the carboxyl-terminated butadiene acrylonitrile copolymer (b) as a liquid polymer but there are no conditions limiting the liquid such as temperature or liquefying the CTBN as a dispersion or solvent solution and that the claimed liquid polymer is not confined to any particular temperature.

The Examiner also argued that (i) the CTBN copolymer of Minamisawa et al. and the Japanese patent are not required to be in any specific form and the Minamisawa's polymer has a Mooney viscosity of 40 to 110 at 100°C, (ii) Minamisawa recognizes that CTBN's are preferably liquid to provide a prepreg of good quality, and (iii) it would be obvious to employ the CTBN of Minamisawa as a liquid in order to facilitate blending with the other liquid components and to provide a prepreg of good quality.

The Examiner also provided a translation of JP 64-60679 and stated that Application Example 5 of JP '670 shows the preparation of an adhesive solution by the same method as that in Application Example 1 where the CTBN is dissolved in a solvent. The Examiner argued that the dissolved CTBN is in a liquid form within the confines of the generally claimed liquid CTBN.

Finally, the Examiner argued that, even if those rationales are not considered, it would have been obvious to use the CTBN's of Minamisawa and the Japanese patent to facilitate processing with the epoxy resin and CTBN-epoxy resin reaction product.

Reply

Contrary to the Examiner's arguments, the claims recite a temperature feature, namely that the reactive liquid polymer (b) is a liquid at ambient temperature. Appellants submit that ambient temperature is a term of art and that a person skilled in the art would recognize the term as referring to the usual

surrounding environmental conditions, e.g., room temperature conditions.³ Even if the claim does not recite particular values, a person skilled in the art would not consider 100°C to be a usual surrounding environmental condition and would not consider that to be ambient temperature. Therefore, the Examiner's argument that Minamisawa's nitrile rubber with a Mooney viscosity between 40 and 110 at 100°C falls within the claimed liquid polymer at ambient temperature is inapposite.

As Appellant's have also previously addressed, Minamisawa's disclosure at col. 3, lines 19-27 also fails to render the claims obvious. Minamisawa may disclose using a liquid CTBN to form the reaction product component (D) for its composition, but it completely fails to suggest using such materials as a separate component. Rather, Minamisawa only discloses using solid nitrile rubbers as its component (E). Given that Minamisawa discloses that a particular component is preferably a liquid but fails to remotely suggest that another component should be a liquid at room temperature, and in fact only discloses using solids for that component, it would not be obvious to modify Minamisawa by employing a reactive liquid polymer as Minamisawa's component (E). Rather, it is only through hindsight in view of Appellant's disclosure that a person skilled in the art would modify Minamisawa to arrive at the present claims.

Additionally, Application Examples 1 and 5 in JP '679 do not teach or suggest that the CTBN is in a liquid form within the confines of the claimed reactive liquid polymer. Example 1 only discloses that all the components are dissolved methyl ethyl ketone to obtain a homogenous solution. There is nothing to suggest that the CTBN is provided as a separate liquid component or that the CTBN is provided as a separate component that by itself is liquid at room temperature.

³ See Exhibit F: Coatings Encyclopedia Dictionary (1995).


CONCLUSION

For the reasons given above, and for the reasons stated in Appellants Brief on Appeal, Appellant respectfully requests that the Honorable Board reverse the Examiner's rejections of claims 1-3, 8-9, 11, and 30.

In the event any fee or additional fee is due in connection with the filing of this paper, the Commissioner is authorized to charge those fees to our Deposit Account No. 18-0988 (Docket No. BFGRP0313USB).

Respectfully submitted,

RENNER, OTTO, BOISSELLE & SKLAR, LLP



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HANDBOOK OF ADHESIVES

Third Edition

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Exhibit E

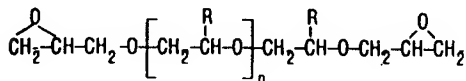
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These high performance resins can be cross-linked with either an aromatic amine or a catalytic curing agent to induce epoxy-to-epoxy homopolymerization. High temperatures are required for these reactions to occur.

Flexible Epoxy Resins

Long chain aliphatic epoxy resins based on a polyglycol or a vegetable oil fatty acid, when reacted with epichlorohydrin, are used as additives to flexibilize epoxy resin adhesives. They are not used alone because of their water sensitivity and overall lack of toughness. They serve as modifiers for Bisphenol A based epoxy resins. An idealized structure for flexible epoxy resins is shown below:



R = AN ORGANIC GROUP OR HYDROGEN

Examples of typical aliphatic epoxy resins based on polyol propylene glycol are D.E.R. 732 and D.E.R. 736 (Dow). Epi-Rez 505 (Interez) and Heloxy WC-85 (Wilmington Chemical) are typical flexible epoxy resins based on a vegetable oil fatty acid.

A compilation of commercial epoxy resins used in adhesives is shown in Table 1.

CURING AGENTS USED IN ADHESIVES

The selection of an appropriate curing agent is as important as selecting the proper epoxy resin. The type of curing agent will determine the rate of reactivity, degree of exotherm, formulation viscosity, gel time, and the heat requirement during the cure cycle. In addition, application technique, pot life requirements, and the desired performance properties of the cured adhesive must be considered when selecting an epoxy curative. Curing agents determine the type of chemical bonds formed and the degree of crosslinking which occur with the epoxy resin. These factors, in turn, affect the chemical resistance, electrical and physical properties, and the heat resistance of the cured adhesive.

The types and number of curing agents available continues to grow rapidly. The chart in Table 2, taken from Dow Chemical's "General Guide to Formulating with Dow Epoxy Resins," summarizes the applications and advantages/disadvantages of different types of curing agents.

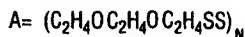
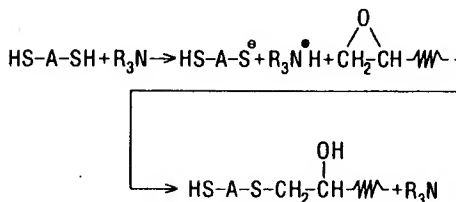
A brief description of various curing agents is provided to aid in the selection of an appropriate curing agent for a variety of adhesive applications.

Polysulfides

An example of a typical polysulfide and its reaction sequence with an epoxide group is shown below:

REACTION SEQUENCE

MERCAPTAN



R = ALIPHATIC
CYCLOALIPHATIC
AROMATIC

The reaction between the terminal mercaptan and epoxide group proceeds very slowly at ambient temperature. However, the introduction of a basic material such as a tertiary amine greatly accelerates the cure. Polysulfides are typically used at ratios of 1:1 or less with epoxy resins, and can be used as co-curing agents with aliphatic amines. Stoichiometric quantities of aliphatic amine and 25 to 50 parts by weight of polysulfide will react with 100 parts by weight of epoxy resin.

Polysulfide/epoxy systems accelerated with tertiary amines exhibit good flexibility and tensile strength at ambient temperature. The aliphatic amine/polysulfide co-curing agent systems yield improved initial elevated tem-

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Table 1. Epoxy Resins Used in Adhesives.

| Type | Viscosity @ 25°C, cps or Durrant's softening point | Epoxide Equivalent Weight | Commercial Products | Remarks |
|----------------------------------|---|---------------------------------|--|--|
| Bisphenol A resins | | | | |
| low viscosity | 4000-6500 | 172-180 | D.E.R. 332 (Dow) Epon 825 (Shell); Araldite GY 6004 (Ciba Geigy); Epo-Tuf 37-15 (Reich- hold) | Virtually pure diglycidyl ether of Bisphenol A |
| medium viscosity | 7000-10,000 | 176-190 | D.E.R. 330, D.E.R. 383, Araldite GY 6008, Epon 826, Epi-Rez 509 (Interez) | Undiluted resins with viscosity lower than standard resins. |
| standard liquid | 11,000-14,000 | 182-195 | D.E.R. 331, Araldite GY 6010, Epon 828, Epi-Rez 510, Epo-Tuf 37-140 | Standard un- diluted epoxy res- ins |
| high viscosity | 16,000-25,000 | 200-250 | D.E.R. 317, D.E.R. 337, Araldite GY 6020, Epon 834, Epo-Tuf 37-141 | |
| solid | 75-85°C, Durrant's softening points | 500-575 | D.E.R. 661, Epon 1001, Epi-Rez 520°C, Epo-Tuf 37-001, Araldite GT 7071 D.E.N. 431 | Low melting solid resin |
| Epoxy novolac resins | 1100-1700 @ 52°C | 172-179 | | Phenolic no- volac |
| | 20,000-50,000 @ 52°C | 176-181 | D.E.N. 438, Ar- aldite EPN 1138, D.E.N. 444 | Phenolic no- volac |
| Epoxy cresol novo- lac resins | 175-350 @ 150°C | 180-220 | Quatrex 3310 (Dow) | High-purity electronic grades |
| | 350-700 @ 150°C | 180-230 | Quatrex 3410 | |
| | 700-1300 @ 150°C | 190-230 | Quatrex 3710 | |
| | 73°C SP | 215-230 | Araldite 1273 | |
| | 80°C SP | 235 Max. | Araldite 1280 | |
| | 99°C SP | 220-245 | Araldite 1299 | |

perature performance. Both systems, however, lose some flexibility on aging.

Amines

The functionality of an amine is determined by the number of amine hydrogens present on the

molecule. A primary amine group (nitrogen with two hydrogens bound to it) will react with two epoxide groups. A secondary amine group (nitrogen with only one hydrogen bound to it) will react with one epoxide group. A tertiary amine group (nitrogen with no hydrogens

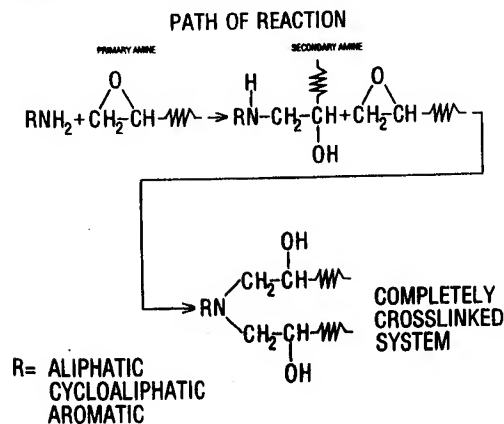
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Table 2. Applications and Characteristics of Epoxy Curing Agents.

| Type | Advantage | Disadvantage | Application |
|-----------------------|--|--|--|
| Polysulfides | Moisture insensitive, quick set time, flexible | Odor, poor elevated temperature performance | Adhesives, sealants |
| Aliphatic amines | Convenience, room temperature cure, low viscosity, low formulation cost | Critical mix ratios, strong skin irritant, high vapor pressure, blushes | Civil engineering, adhesives, grouts, casting and electrical encapsulation |
| Polyamides | Convenience, room temperature cure, low toxicity, good flexibility or resilience, good toughness | Higher formulation cost, high viscosity, low heat resistance, low vapor pressure | Civil engineering, adhesives, grouts, castings, coatings |
| Amidoamines | Reduced volatility, convenient mix ratios, good toughness | Poor elevated temperature performance, some incompatibility with epoxy resin | Construction adhesives, concrete bonding, troweling compounds |
| Aromatic amines | Moderate heat resistance, good chemical resistance | Solids at room temperature, long elevated-temperature cure schedules | Filament wound pipe, electrical encapsulation, <u>adhesives</u> |
| Dicyandiamide | Latent cure, good elevated temperature properties, good electrical properties | Long elevated-temperature cure, insoluble in resin | Powder coatings, electrical laminates, one-component adhesives |
| Catalytic | Extremely long pot life, high heat resistance | Long elevated-temperature cure schedules, poor moisture resistance | Adhesives, electrical, encapsulation, powder coatings, electric laminates |
| Anhydrides | Good heat resistance, good chemical resistance | Long elevated-temperature cure schedules, critical mix ratios | Filament wound pipe, electrical encapsulation and adhesives |
| Melamine/formaldehyde | Good hardness and flexibility, one-package stability, solvent-free systems | Elevated temperature cure | Waterborne coatings, container and deco finishes |
| Urea/formaldehyde | Good film color, one-package stability, good intercoat adhesion | Elevated temperature cure | Fast bake enamels, primers and topcoats |
| Phenol/formaldehyde | Good elevated temperature properties, good chemical resistance, good hardness and flexibility | Solid, poor weatherability | Powder coatings, molding compounds |

bound to it) will not react readily with an epoxide group, but will act as a catalyst to accelerate epoxy reactions. A typical amine reaction sequence is shown below:

Theoretically, the hydroxyl groups generated in the above reaction sequence are capable of reacting with other epoxide groups, thus forming an ether linkage. This reaction can be catalyzed by tertiary amines.



Aliphatic Amines

The liquid aliphatic polyamines and their adducts are convenient to handle and give good cured resin physical properties, including excellent chemical and solvent resistance. Mix ratios are critical for optimum performance. Aliphatic amines offer fast curing at ambient temperatures. Pot life is short and high exotherm in thick sections or large masses can lead to thermal decomposition. Good long-term retention of properties is possible at temperatures

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up to 100°C (212°F). Short-term exposure to higher temperatures can be tolerated. Epoxy resin formulations containing aliphatic amines will blush under very humid conditions.

Adducted aliphatic polyamines such as D. E. H. 52 (Dow) epoxy hardener and D. E. H. 58 (Dow) epoxy hardener offer the advantages of lower vapor pressure, reduced tendency to blush, and less critical mix ratios.

Cycloaliphatic Amines

When compared to aliphatic polyamines, cycloaliphatic amines produce cured resins having improved thermal resistance and toughness. Glass transition temperatures approach those of aromatic amines while percent elongation can be doubled. Because cycloaliphatic amines are less reactive than aliphatic polyamines, their use results in a longer pot life and in the ability to cast larger masses.

Aromatic Amines

Aromatic amines are solids at room temperature. These hardeners are routinely melted at elevated temperatures and blended with warmed resin. Eutectic mixtures of metaphenylene diamine and methylene dianiline exhibit a depressed melting point resulting in an aromatic hardener that remains a liquid over short periods of time. The pot life is considerably longer than that achieved with aliphatic polyamines. Elevated temperature cures are required to develop optimum properties which are maintained at temperatures up to 150°C (300°F). Aromatic amines have improved chemical and thermal resistance when compared to aliphatic polyamines.

Polyamides

The most commonly used polyamides are the condensation products of dimerized fatty acids and aliphatic amines such as diethylene triamine. A range of molecular weights are available making polyamide curing agents versatile in a variety of adhesive applications. The polyamides react with epoxide groups through the unreacted amine functional groups in the polyamide backbone. As a result of their relatively large molecular weight, the ratio of

polyamide to epoxy is more forgiving (less critical) than with low molecular weight polyamines. Polyamides also offer the advantages of curing without blushing, and improved adhesion. However, they are much darker in color than polyamines.

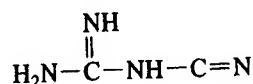
The various molecular weight polyamides exhibit different degrees of compatibility with epoxy resins. To ensure optimum properties, the polyamide/epoxy mixture must be allowed to react partially before being used. The partial reaction assures compatibility and is known as the induction period. Because polyamides have a long pot life, the induction time does not significantly shorten the usable time of the system.

Polyamide-cured epoxides lose structural strength rapidly with increasing temperature. This limits their use to adhesive applications which will not be subjected to temperatures above 65°C (148°F).

Amidoamines

Amidoamines are derivatives of monobasic carboxylic acids (such as ricinoleic acid) and an aliphatic polyamine. Like the polyamides, amidoamines can be used over a range of additive levels to enhance a specific property. The reactivity of amidoamines with epoxies is similar to that of polyamides. However, amidoamines offer several advantages over aliphatic amines and polyamines: more convenient mix ratios, increased flexibility, better moisture resistance than aliphatic polyamines, and lower viscosity and color than polyamides.

Dicyandiamide



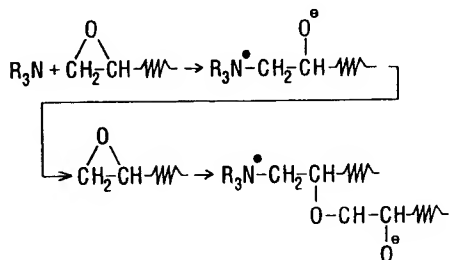
Dicyandiamide (Dicy) is a solid curing agent which, when ball milled into liquid epoxy resins, provides one-package stability for up to six months at ambient temperature. Cures occur with heating to 150°C (300°F). A tertiary amine accelerator is necessary for rapid cures. Dicy offer the advantage of being latent (reacts with epoxy resins upon heating, and stops reacting when the heat is removed). This par-

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tially cured or "B-staged" state is ideal for prepregs for supported film adhesives. Typically, dicyandiamide is used at levels of 5-7 parts per 100 parts liquid epoxy resin, and at levels of 3-4 parts per 100 parts solid epoxy resin.

Catalytic Curing Agents

Catalytic curing agents are a group of compounds which promote epoxy to epoxy reactions without being consumed in the process. A typical epoxy homopolymerization using a tertiary amine is shown below:



Stable, one-package systems can be developed with many catalytic curing agents such as the boron trifluoride complexes. Tertiary amines and amine salts have pot lives generally ranging from 2 to 24 hours. The latent catalysts are activated by heat and cause a disassociation of the active catalyst from the blocking group.

The amount of catalyst used may vary from 2 to 10 parts per 100 parts resin. To determine the best ratio of catalyst to resin, several different catalyst levels should be evaluated to determine which level provides maximum properties. Several common catalytic curing agents are benzyldimethylamine (BDMA), boron trifluoride monoethylamine ($\text{BF}_3 \cdot \text{MEA}$), and 2-methylimidazole (2-MI).

Anhydrides

Liquid and solid anhydrides are used extensively to cure epoxy resins. The reactivity of some anhydrides with epoxies is slow, therefore an accelerator, usually a tertiary amine, is often used at 0.5–3% to speed gel time and cure. The optimum amount is usually critical, and is dependent on the anhydride, the resin used, and cure schedules. The amount of accelerator above or below the "optimum" concentration can result in reduced high

temperature performance. The optimum concentration of accelerator should be determined experimentally. Eutectic mixtures to depress melting points may be prepared.

The reaction of anhydrides with epoxide groups is complex, with several competing reactions capable of taking place. The three most important are:

1. The opening of the anhydride ring with an alcoholic hydroxyl to form the monoester.
2. Subsequent to the opening of the ring, the nascent carboxylic groups react with the epoxide to give an ester linkage.
3. The epoxide groups react with nascent or existing hydroxyl groups, catalyzed by the acid, producing an ether linkage.

At low elevated temperature cures, the ether and ester reactions take place at about the same frequency. At higher temperatures, the ester linkage occurs more frequently and this probably accounts for the reduced elevated temperature performance of systems gelled at initially high temperatures. Because reaction 3 can take place independently in the acid medium, the ratio of anhydride to epoxy is less critical than with an amine. It can vary from 0.5 to 0.9 equivalents of epoxy and is determined experimentally to achieve desired properties.

Compared to aliphatic amine cures, the pot life of epoxy-anhydride formulations is usually long and exotherm is low. Elevated temperature cures (up to 200°C) are necessary and long post-cures are required to develop ultimate properties. Electrical and physical strength properties are good over a wide temperature range. Compared to amine-cured systems, anhydride-cured systems offer better chemical resistance to aqueous acids and less chemical resistance to some reagents.

SUMMARY

The most commonly used curing agents for ambient cured adhesives are polyamides, aliphatic amines, cycloaliphatic amines and amidoamines. The choice of hardener is dependent on the performance requirements of the adhesive. For high performance, high temperature

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Table 3. Curing Agents for Epoxy Resins Adhesives.

| <i>Curing agent</i> | <i>Recommended Parts/100 parts Liquid resin</i> | <i>Curing Temperature °C</i> | <i>Curing agent Suppliers and Trade Names</i> |
|---|---|--------------------------------------|--|
| Aliphatic amines | | | |
| diethylene triamine (DETA) | 8-10 | R.T.-150 | D.E.H. (The Dow Chemical Company) |
| triethylene tetramine (TETA) | 10-13 | R.T.-150 | Amicure (Pacific Anchor Chemical Co.) |
| Aminoethyl piperazine (AEP) | 20-23 | R.T.-150 | Epo-Tuf (Reichhold Chemical) |
| Aromatic amines | | | |
| methylene dianiline | 51-55 | 175 (2 hr) | Curithane (Dow) |
| 4,4-diaminodiphenyl sulfone | 30-34 | 175 (2 hr) | |
| MDA/MPDA eutectics | | | Amicure (Air Products and Chemicals, Inc.) Ancamine (Pacific Anchor) Epon (Shell Chemical Company) |
| Cycloaliphatic amines | | | |
| wide variety of modified products from major curing agent suppliers | | R.T.-150 | Amicure (Air Products) Ancamine (Pacific Anchor) Azamine (SHEREX) Epo-Tuf (Reichhold) Versamine (Henkel Corporation) |
| Polyamides | | | |
| wide variety of polyamide curing agent with range of molecular weights. | | R.T. to 2 hr @ 100 | All major curing agent suppliers listed above offer Polyamide curing agents. |
| Heat activated curing agents and catalysts | | | |
| benzyl dimethylamine (BDMA) | 2-4 | 150 (2 hr) | Air Products and Chemical, Inc. |
| dicyandiamide (DICY) | 2-4 | 150 (2 hr) | Pacific Anchor Chemical Corp. |
| boron trifluoride amine complexes | 2-4 | 150 (2 hr) | Allied Chemical, Ciba-Geigy, Sylvachem (Sub. of Arizona Chem.) |

resistant adhesives, the aromatic amines, diacyandiamide, and catalytic curing agents are used.

Table 3 contains a list of curing agents used in adhesives and their manufacturers.

DILUENTS

Diluents are used in epoxy adhesive formulations to reduce viscosity and to allow the use

of large amounts of filler. Both reactive and nonreactive diluents are used. Reactive diluents are low viscosity mono or difunctional epoxy materials. Table 4 lists reactive epoxy diluents in descending order of viscosity.

Nonreactive diluents are not as widely used as reactive diluents in epoxy resin adhesives because they decrease the cured properties more than a reactive diluent and have a tendency to migrate to the surface of the adhesive bond. The

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aluminum powder

aluminum powder POWDERED ALUMINUM METAL used as a PIGMENT. Available in LEAFING or NONLEAFING forms.⁴¹

Density, 2.73 g/cm³ (22.5 lb/gal).

ASTM Test Method D480 covers procedures for sampling, qualitative analysis, and physical testing of flaked aluminum powders and pastes (leafing and nonleafing) for coatings.

aluminum primer (1) PRIMER specifically formulated for aluminum metal; (2) primer containing a proportion of aluminum PIGMENT, but distinguished from aluminum paints in which the aluminum is designed to float to the top of the film giving metallic brilliance, a feature undesirable in a primer.^{71,158} (BSI)

Aluminum primers have been used on resinous TIMBER or timber which has been treated with oil-soluble wood preservatives.

aluminum resinate brown soft mass, insoluble in water; used for water-repellancy and as a DRIER in VARNISH.⁸³

aluminum silicate (clay) Al₂O₃·2SiO₂·2H₂O. Pigment White 19 (77005). White inert PIGMENT of little color and opacity, obtained from certain natural deposits of CHINA CLAY, KAOLIN, FELDSPAR, and similar materials.^{41,58} Syn: kaolin, hydrated aluminum silicate, china clay, pipe clay, white bole

Hydrated aluminum silicate, fine grain crystallized clay. Laminar structure with repeating alumina-silica configurations. Density, 2.58 g/cm³ (21.5 lb/gal); O.A., 32.1-55 lb/100 gal. Particle size, 0.5-3.5 µm.

aluminum soaps aluminum compounds of FATTY ACIDS and naphthenic acid, etc.¹²⁷

These materials are soluble generally in paint vehicles and THINNERS, and are employed as THICKENING, FLATTING, and WATERPROOFING agents.

aluminum stearate complex SALT of SOAP of ALUMINUM and STEARIC ACID. It is a white powder, which forms COLLOIDAL SOLUTIONS or gels with DRYING OILS and certain SOLVENTS.^{58,83,139}

It is used as a FLATTING AGENT, ANTI-SETTLING AGENT for pigments, and to help prevent penetration.

amber a fossil RESIN found chiefly in the blue earth of East Prussia. It is primarily an exudation of *Pinus succinifera*.¹⁶⁷

ambient usual surrounding environmental conditions, such as pressure or temperature.

ambient air the outdoor air in a particular location.^{171,172} (IUPAC)

ambient air quality a general term used to describe the quality of the outside air.^{171,172} (IUPAC)
Usually adjectives such as good, fair, bad, etc. are used by the media to describe this; often some

form of AIR POLLUTION or air quality index is employed to determine the specific descriptive term to be used. These are very qualitative terms of little or no scientific value. (IUPAC)

ambient cure see SELF-CURING.^{67,55}

American Chemical Society, The (ACS) is a nonprofit scientific and educational association of professional chemists and chemical engineers. Founded in New York City on April 6, 1876, with 133 charter members, the Society now has a membership of over 110,000 and is the world's largest organization devoted to a single science. It carries out a broad program of publications, meetings, and other activities in pursuit of the following objectives, as set forth in its National Charter granted by Congress in 1937: to encourage in the broadest and most liberal manner the advancement of chemistry in all its branches; the promotion of research in chemical science and industry; the improvement of the qualifications and usefulness of chemists through high standards of professional ethics, education, and attainments; the increase and diffusion of chemical knowledge; and by its meetings, professional contacts, reports, papers, discussions, and publications, to promote scientific interests and inquiry; thereby fostering public welfare and education, aiding the development of our country's industries, and adding to the material prosperity and happiness of our people.^{144,174}

Its address is 1155 Sixteenth Street, N.W., Washington, D.C. 20036; Tel: (202) 872-4600.

American Conference of Governmental Industrial Hygienists (ACGIH) an organization of professionals in governmental agencies or educational institutions engaged in occupational safety and health programs. ACGIH develops and publishes recommended occupational exposure limits for chemical substances and physical agents.¹⁷⁴ See TLV.¹⁷²

Its address is 6500 Glenway Ave., Bldg. D-7, Cincinnati, OH 45211; Tel.: (513) 661-7881.

American gallon see gallon, U.S.¹²⁸

American National Standards Institute (ANSI) a privately funded organization that identifies industrial/public national consensus standards and coordinates their development. Many ANSI standards relate to safe design/performance of equipment and safe practices or procedures.¹⁷⁴

Its address is: 1430 Broadway, New York, NY 10018; Tel.: (212) 642-4900.

American process zinc oxide zinc oxide PIGMENT made directly from zinc ores.^{41,131} See ZINC OXIDE.⁴¹

Sometimes called "direct" process.

Words presented in CAP/SMALL CAPS type indicate that the word is defined in another part of the *Coatings Encyclopedic Dictionary*. Numerical superscripts classify terms in one or more of the categories listed in the second section of this volume.